Evaluation of vorticity expansion approximation of current-density functional theory by means of Levy's asymptotic bound

Masahiko Higuchi
Department of Physics, Faculty of Science, Shinshu University, Matsumoto 390-8621, Japan

Katsuhiko Higuchi
Graduate School of Advanced Sciences of Matter, Hiroshima University, Higashi-Hiroshima 739-8527, Japan
(Received 27 October 2006; revised manuscript received 9 February 2007; published 14 May 2007)

We evaluate the vorticity expansion approximation (VEA) of the current-density functional theory (CDFT) by means of the derived Levy's asymptotic bound. The VEA formula exactly satisfies Levy's asymptotic bound, though the local-density approximation of the CDFT does not. The validity of the VEA formula is thus confirmed successfully.

DOI: 10.1103/PhysRevB.75.195114 PACS number(s): 71.15.Mb, 31.15.Ew, 31.25.Eb

I. INTRODUCTION

The density functional theory (DFT) provides a powerful and reliable approach to the electronic structures of many-electron systems.[1,2] The DFT has been extended so that various physical quantities can be chosen as basic variables.[3–5] For instance, the spin-density functional theory,[6,7] the current-density functional theory (CDFT),[8–11] and LDA+U method[12–15] are regarded as examples of such extensions.

In the CDFT, we choose the paramagnetic current density as a basic variable in addition to the electron density. Therefore, the CDFT is useful for describing the ground-state properties of systems such as open shell atoms, inhomogeneous electronic systems in an external magnetic field, and f-electron materials where an orbital current is induced from both the strong spin-orbit interaction and the intra-atomic Coulomb interaction.

For the practical use of the CDFT, one has to develop the approximate form of the exchange-correlation energy functional. Many attempts to develop the approximate form have been presented so far.[8,9,16–29] We have already suggested two strategies for treating the approximate form.[22] One is to start with the coupling-constant expression of the exchange-correlation energy functional of the CDFT.[22] By using this expression, the local-density approximation (LDA), average-density approximation, and weighted-density approximation have been proposed similarly to those in the conventional DFT.[22] Another strategy is to utilize as constraints exact relations that are satisfied with the exchange and correlation energy functionals.[22] This strategy is analogous to the one used for developing the generalized gradient approximation[30–32] in the conventional DFT. To date, many exact relations have been derived by means of the virial theorem, uniform and nonuniform scaling properties.[18,19,22,25,28]

Along the latter strategy, we have recently proposed the vorticity expansion approximation (VEA) for the exchange and correlation energy functionals.[29] Many exact relations that are derived from uniform and nonuniform scalings of electron coordinates are satisfied with the VEA formulas.[29] Due to the well-behaved forms, the VEA formulas can well reproduce the exchange and correlation energies of the homogeneous electron liquid under a uniform magnetic field.[29] However, Levy's asymptotic bound,[33,34] which is regarded as an important constraint and has been very effectively used in developing the Perdew-Burke-Ernzerhof (PBE) functional of the conventional DFT,[32] has not been taken into account in developing the VEA of the CDFT.[29]

Levy's asymptotic bound is such that the correlation energy functional for the uniformly scaled density approaches a constant in the limit of infinite scaling factor.[33,34] This bound is not satisfied with the LDA of the conventional DFT because the short-range correlation term[35,36] causes the logarithmic divergence in the limit of infinite scaling factor. The additional term that expresses the effect of the density gradient was devised in the PBE functional so as to avoid such divergence.[32] Thus, this bound has played an important role in the development of the PBE functional. However, this bound has not yet been proven for the correlation energy functional of the CDFT. Also in the CDFT, this exact property would be true and very useful for developing, modifying, or evaluating the approximate forms of the exchange and correlation energy functionals.

In this paper, we derive Levy's asymptotic bound in the framework of the CDFT and evaluate both the VEA and LDA formulas. The organization of this paper is as follows. In Sec. II, we shall give a proof of Levy's asymptotic bound for the correlation energy functional of the CDFT. In Secs. III and IV, it is shown that Levy's asymptotic bound is satisfied with the VEA correlation energy functional while it is not with the CDFT-LDA one. Finally, a conclusion is given in Sec. V.

II. LEVY'S ASYMPTOTIC BOUND FOR THE CORRELATION ENERGY FUNCTIONAL OF THE CDFT

In this section, we derive Levy's asymptotic bound for the correlation energy functional of the CDFT. Let us start with the definition of the correlation energy functional $E_c[\rho,\mathbf{j}_p]$, where $\rho(\mathbf{r})$ and $\mathbf{j}_p(\mathbf{r})$ denote the electron density and the paramagnetic current density, respectively. According to the constrained-search formulation of the CDFT, $E_c[\rho,\mathbf{j}_p]$ is defined by[18,22,29]

$$E_c[\rho,\mathbf{j}_p] \coloneqq \langle\Psi[\rho,\mathbf{j}_p]|\hat{T}+\hat{W}|\Psi[\rho,\mathbf{j}_p]\rangle - \langle\Phi[\rho,\mathbf{j}_p]|\hat{T}+\hat{W}|\Phi[\rho,\mathbf{j}_p]\rangle, \quad (1)$$

where $\hat{T}$ and $\hat{W}$ are operators of the kinetic energy and the electron-electron interaction, respectively. $\Psi[\rho,\mathbf{j}_p]$ is the wave function which minimizes the expectation value of $\hat{T}+\hat{W}$ in the constrained search, i.e.,

$$F[\rho,\mathbf{j}_p] \coloneqq \min_{\Psi\to(\rho,\mathbf{j}_p)} \langle\Psi|\hat{T}+\hat{W}|\Psi\rangle =: \langle\Psi[\rho,\mathbf{j}_p]|\hat{T}+\hat{W}|\Psi[\rho,\mathbf{j}_p]\rangle, \quad (2)$$

and $\Phi[\rho,\mathbf{j}_p]$ is the minimizing single Slater determinant, which is defined in the constrained search of the kinetic-energy functional,

$$T_s[\rho,\mathbf{j}_p] \coloneqq \min_{\Phi\to(\rho,\mathbf{j}_p)} \langle\Phi|\hat{T}|\Phi\rangle =: \langle\Phi[\rho,\mathbf{j}_p]|\hat{T}|\Phi[\rho,\mathbf{j}_p]\rangle. \quad (3)$$

From Eq. (3), the minimizing single Slater determinant $\Phi[\rho,\mathbf{j}_p]$ satisfies

$$\left[\hat{T}+\int v_s(\mathbf{r})\hat{\rho}(\mathbf{r})d\mathbf{r}+\int \mathbf{A}_s(\mathbf{r})\cdot\hat{\mathbf{j}}_p(\mathbf{r})d\mathbf{r}\right]\Phi[\rho,\mathbf{j}_p](\mathbf{r}_1,\ldots,\mathbf{r}_N) = E_s\Phi[\rho,\mathbf{j}_p](\mathbf{r}_1,\ldots,\mathbf{r}_N), \quad (4)$$

where E_s, $v_s(\mathbf{r})$, and $\mathbf{A}_s(\mathbf{r})$ are the Lagrange multipliers which correspond to constraints

$$\langle\Phi[\rho,\mathbf{j}_p]|\Phi[\rho,\mathbf{j}_p]\rangle = 1, \quad (5)$$

$$\langle\Phi[\rho,\mathbf{j}_p]|\hat{\rho}(\mathbf{r})|\Phi[\rho,\mathbf{j}_p]\rangle = \rho(\mathbf{r}), \quad (6)$$

and

$$\langle\Phi[\rho,\mathbf{j}_p]|\hat{\mathbf{j}}_p(\mathbf{r})|\Phi[\rho,\mathbf{j}_p]\rangle = \mathbf{j}_p(\mathbf{r}), \quad (7)$$

respectively. Here, $\hat{\rho}(\mathbf{r})$ and $\hat{\mathbf{j}}_p(\mathbf{r})$ stand for operators of the electron density and the paramagnetic current density, respectively.

With reference to the derivation procedure of the original Levy's asymptotic bound,[33] we consider the following equation:

$$\left[\hat{T}+\int v_s(\mathbf{r})\hat{\rho}(\mathbf{r})d\mathbf{r}+\int \mathbf{A}_s(\mathbf{r})\cdot\hat{\mathbf{j}}_p(\mathbf{r})d\mathbf{r}+\alpha\hat{W}\right]\times\Psi_s^\alpha(\mathbf{r}_1,\ldots,\mathbf{r}_N) = E_s^\alpha\Psi_s^\alpha(\mathbf{r}_1,\ldots,\mathbf{r}_N), \quad (8)$$

where α is a coupling constant of the electron-electron interaction. Ψ_s^α and E_s^α are supposed to be the ground-state wave function and its eigenvalue, respectively. Transforming Eq. (8) by the coordinate scaling of electrons such that $\mathbf{r}_i \to \lambda^{-1}\mathbf{r}_i$, we get

$$\left[\hat{T}+\lambda^2\int v_s(\lambda\mathbf{r})\hat{\rho}(\mathbf{r})d\mathbf{r}+\lambda\int \mathbf{A}_s(\lambda\mathbf{r})\cdot\hat{\mathbf{j}}_p(\mathbf{r})d\mathbf{r}+\alpha\lambda\hat{W}\right]\times\lambda^{3N/2}\Psi_s^\alpha(\lambda\mathbf{r}_1,\ldots,\lambda\mathbf{r}_N) = \lambda^2E_s^\alpha\lambda^{3N/2}\Psi_s^\alpha(\lambda\mathbf{r}_1,\ldots,\lambda\mathbf{r}_N), \quad (9)$$

where $\lambda^{3N/2}\Psi_s^\alpha(\lambda\mathbf{r}_1,\ldots,\lambda\mathbf{r}_N)$ is a scaled wave function with the normalizing constant $\lambda^{3N/2}$. In the above transformation, we used the explicit forms of $\hat{T}$, $\hat{W}$, $\hat{\rho}(\mathbf{r})$, and $\hat{\mathbf{j}}_p(\mathbf{r})$, which are given by $\hat{T}=\Sigma_{i=1}^N\frac{\mathbf{p}_i^2}{2m}$, $\hat{W}=\frac{e^2}{2}\Sigma_{i=1}^N\Sigma_{j\neq i}^N\frac{1}{|\mathbf{r}_i-\mathbf{r}_j|}$, $\hat{\rho}(\mathbf{r})=\Sigma_{i=1}^N\delta(\mathbf{r}-\mathbf{r}_i)$, and $\hat{\mathbf{j}}_p(\mathbf{r})=\frac{1}{2m}\Sigma_{i=1}^N[\mathbf{p}_i\delta(\mathbf{r}-\mathbf{r}_i)+\delta(\mathbf{r}-\mathbf{r}_i)\mathbf{p}_i]$, respectively. If α is set to be equal to λ^{-1}, then Eq. (9) is rewritten as

$$\hat{H}_\lambda\lambda^{3N/2}\Psi_s^{\lambda^{-1}}(\lambda\mathbf{r}_1,\ldots,\lambda\mathbf{r}_N) = \lambda^2E_s^{\lambda^{-1}}\lambda^{3N/2}\Psi_s^{\lambda^{-1}}(\lambda\mathbf{r}_1,\ldots,\lambda\mathbf{r}_N) \quad (10)$$

with

$$\hat{H}_\lambda \coloneqq \hat{T}+\hat{W}+\lambda^2\int v_s(\lambda\mathbf{r})\hat{\rho}(\mathbf{r})d\mathbf{r}+\lambda\int \mathbf{A}_s(\lambda\mathbf{r})\cdot\hat{\mathbf{j}}_p(\mathbf{r})d\mathbf{r}. \quad (11)$$

It should be noted that $\lambda^{3N/2}\Psi_s^{\lambda^{-1}}(\lambda\mathbf{r}_1,\ldots,\lambda\mathbf{r}_N)$ is the ground-state wave function of $\hat{H}_\lambda$. The proof is given in Appendix A.

In order to derive Levy's asymptotic bound, we shall consider the correlation energy functional for the scaled basic variables $\rho_\lambda(\mathbf{r})$ and $\mathbf{j}_{p\lambda}(\mathbf{r})$ which are given by[18]

$$\rho_\lambda(\mathbf{r}) = \lambda^3\rho(\lambda\mathbf{r}) \quad (12)$$

and

$$\mathbf{j}_{p\lambda}(\mathbf{r}) = \lambda^4\mathbf{j}_p(\lambda\mathbf{r}), \quad (13)$$

respectively. From definition (1), the correlation energy functional for the scaled basic variables is written by

$$\begin{aligned} E_c[\rho_\lambda,\mathbf{j}_{p\lambda}] &= \langle\Psi[\rho_\lambda,\mathbf{j}_{p\lambda}]|\hat{T}+\hat{W}|\Psi[\rho_\lambda,\mathbf{j}_{p\lambda}]\rangle - \langle\Phi[\rho_\lambda,\mathbf{j}_{p\lambda}]|\hat{T}+\hat{W}|\Phi[\rho_\lambda,\mathbf{j}_{p\lambda}]\rangle \\ &= \langle\Psi[\rho_\lambda,\mathbf{j}_{p\lambda}]|\hat{H}_\lambda|\Psi[\rho_\lambda,\mathbf{j}_{p\lambda}]\rangle - \langle\Phi[\rho_\lambda,\mathbf{j}_{p\lambda}]|\hat{H}_\lambda|\Phi[\rho_\lambda,\mathbf{j}_{p\lambda}]\rangle. \end{aligned} \quad (14)$$

In the second line, we use the fact that $\Psi[\rho_\lambda,\mathbf{j}_{p\lambda}]$ and $\Phi[\rho_\lambda,\mathbf{j}_{p\lambda}]$ yield the same scaled basic variables. Since $\lambda^{3N/2}\Psi_s^{\lambda^{-1}}(\lambda\mathbf{r}_1,\ldots,\lambda\mathbf{r}_1)$ is the ground-state wave function of $\hat{H}_\lambda$, as is shown in Appendix A, the following inequality holds:

$$\langle\Psi[\rho_\lambda,\mathbf{j}_{p\lambda}]|\hat{H}_\lambda|\Psi[\rho_\lambda,\mathbf{j}_{p\lambda}]\rangle \geqslant \langle\lambda^{3N/2}\Psi_s^{\lambda^{-1}}|\hat{H}_\lambda|\lambda^{3N/2}\Psi_s^{\lambda^{-1}}\rangle = \lambda^2E_s^{\lambda^{-1}}. \quad (15)$$

Substituting this inequality into Eq. (14), we get

$$E_c[\rho_\lambda,\mathbf{j}_{p\lambda}] \geqslant \lambda^2E_s^{\lambda^{-1}} - \langle\Phi[\rho_\lambda,\mathbf{j}_{p\lambda}]|\hat{H}_\lambda|\Phi[\rho_\lambda,\mathbf{j}_{p\lambda}]\rangle. \quad (16)$$

Let us consider the first term on the right-hand side of Eq. (16). If α is very small in Eq. (8), then the last term of Eq.

(8) can be regarded as the perturbation. Then, one can express E_s^α in the form of the perturbation expansion

$$E_s^\alpha = E_0 + \alpha E_1 + \alpha^2 E_2 + O(\alpha^3), \tag{17}$$

where E_0, E_1, and E_2 denote the unperturbed, the first-, and second-order perturbation energies, respectively. Further, Eq. (17) is rewritten as

$$\lambda^2 E_s^{\lambda^{-1}} = \lambda^2 E_0 + \lambda E_1 + E_2 + O(\lambda^{-1}). \tag{18}$$

Comparing Eq. (4) with Eq. (8), $\Phi[\rho,\mathbf{j}_p]$ can be regarded as the unperturbed wave function. Thus, E_0 and E_1 are given by

$$E_0 = \langle \Phi[\rho,\mathbf{j}_p]|\hat{T} + \int v_s(\mathbf{r})\hat{\rho}(\mathbf{r})d\mathbf{r} + \int \mathbf{A}_s(\mathbf{r})\cdot\hat{\mathbf{j}}_p(\mathbf{r})d\mathbf{r}|\Phi[\rho,\mathbf{j}_p]\rangle = E_s^0 \tag{19}$$

and

$$E_1 = \langle \Phi[\rho,\mathbf{j}_p]|\hat{W}|\Phi[\rho,\mathbf{j}_p]\rangle, \tag{20}$$

respectively.

Next, let us consider the second term on the right-hand side of Eq. (16). It is known that the minimizing wave function $\Phi[\rho,\mathbf{j}_p]$ has the following property:[25]

$$\Phi[\rho_\lambda,\mathbf{j}_{p\lambda}](\mathbf{r}_1,\ldots,\mathbf{r}_N) = \lambda^{3N/2}\Phi[\rho,\mathbf{j}_p](\lambda\mathbf{r}_1,\ldots,\lambda\mathbf{r}_N). \tag{21}$$

The equation for $\lambda^{3N/2}\Phi[\rho,\mathbf{j}_p](\lambda\mathbf{r}_1,\ldots,\lambda\mathbf{r}_N)$ can be obtained by setting α to be equal to 0 in Eq. (9). Then, using Eqs. (11) and (21), we get the equation for $\Phi[\rho_\lambda,\mathbf{j}_{p\lambda}]$,

$$(\hat{H}_\lambda - \hat{W})\Phi[\rho_\lambda,\mathbf{j}_{p\lambda}](\mathbf{r}_1,\ldots,\mathbf{r}_N) = \lambda^2 E_s^0\Phi[\rho_\lambda,\mathbf{j}_{p\lambda}](\mathbf{r}_1,\ldots,\mathbf{r}_N). \tag{22}$$

Therefore, the second term on the right-hand side of Eq. (16) is rewritten as

$$\langle \Phi[\rho_\lambda,\mathbf{j}_{p\lambda}]|\hat{H}_\lambda|\Phi[\rho_\lambda,\mathbf{j}_{p\lambda}]\rangle = \lambda^2 E_s^0 + \langle \Phi[\rho_\lambda,\mathbf{j}_{p\lambda}]|\hat{W}|\Phi[\rho_\lambda,\mathbf{j}_{p\lambda}]\rangle = \lambda^2 E_0 + \lambda E_1. \tag{23}$$

Here, Eqs. (19)–(21) and the transformation of the integration variables are used to obtain the second equality.

Substituting Eqs. (18) and (23) into Eq. (16), we obtain

$$E_c[\rho_\lambda,\mathbf{j}_{p\lambda}] \geqslant E_2 + O(\lambda^{-1}). \tag{24}$$

This inequality leads to

$$\lim_{\lambda\to\infty} E_c[\rho_\lambda,\mathbf{j}_{p\lambda}] \geqslant E_2. \tag{25}$$

In addition, we know that $E_c[\rho_\lambda,\mathbf{j}_{p\lambda}] \leqslant 0$ for all λ from definition (1). Therefore, the value of $\lim_{\lambda\to\infty} E_c[\rho_\lambda,\mathbf{j}_{p\lambda}]$ is less than zero. Consequently, $\lim_{\lambda\to\infty} E_c[\rho_\lambda,\mathbf{j}_{p\lambda}]$ takes a finite value, i.e.,

$$\lim_{\lambda\to\infty} E_c[\rho_\lambda,\mathbf{j}_{p\lambda}] = \text{const}. \tag{26}$$

The exchange and correlation energy functionals depend on $\mathbf{j}_p(\mathbf{r})$ only through the vorticity[9,29]

$$\boldsymbol{\nu}(\mathbf{r}) := \nabla\times[\mathbf{j}_p(\mathbf{r})/\rho(\mathbf{r})], \tag{27}$$

so $E_c[\rho,\mathbf{j}_p]$ can be rewritten as a functional of $\rho(\mathbf{r})$ and $\boldsymbol{\nu}(\mathbf{r})$. If we define $\bar{E}_c[\rho,\boldsymbol{\nu}]$ by $\bar{E}_c[\rho,\boldsymbol{\nu}] := E_c[\rho,\mathbf{j}_p]$, then Eq. (26) is rewritten by

$$\lim_{\lambda\to\infty} \bar{E}_c[\rho_\lambda,\boldsymbol{\nu}_\lambda] = \text{const}, \tag{28}$$

with

$$\boldsymbol{\nu}_\lambda(\mathbf{r}) := \nabla\times[\mathbf{j}_{p\lambda}(\mathbf{r})/\rho_\lambda(\mathbf{r})] = \lambda^2\boldsymbol{\nu}(\lambda\mathbf{r}). \tag{29}$$

Equation (26) or (28) is regarded as Levy's asymptotic bound of the CDFT.

III. EVALUATION OF THE VEA FORMULA

In this section, we first give the outline of the VEA of the CDFT, which has been recently developed by us.[29] Then, we shall confirm that such a VEA formula of the correlation energy satisfies Levy's asymptotic bound of the CDFT.

A. VEA formulas of the exchange and correlation energy functionals of the CDFT

In recent years, we have proposed the practical form of the exchange-correlation energy functional of the CDFT.[29] Both exchange and correlation energies are expanded in terms of the vorticity of the paramagnetic current density, which is defined as Eq. (27). The expansions are up to the second order of the vorticity, and the expansion coefficients are determined by requiring them to satisfy sum rules and bounds that have been derived by several workers.[9,18,22] Resultant VEA formula of the exchange energy functional is given by[29]

$$\bar{E}_x[\rho,\boldsymbol{\nu}] = E_x[\rho] + \int \rho(\mathbf{r})|\boldsymbol{\nu}(\mathbf{r})|^2\, D(\rho)|_{\rho=\rho(\mathbf{r})}d\mathbf{r}, \tag{30}$$

where $E_x[\rho]$ is the exchange energy functional of the conventional DFT. $D(\rho)$ is expressed as the power of ρ, i.e.,

$$D(\rho) = \frac{\bar{D}_x\hbar^2}{a_H^3\varepsilon_H}\rho^{-1}, \tag{31}$$

where $\bar{D}_x$ is the dimensionless constant, and a_H and ε_H stand for the Bohr radius and Rydberg constant, respectively.

Also, the VEA formula of the correlation energy functional is given by[29]

$$\bar{E}_c[\rho,\boldsymbol{\nu}] = E_c[\rho] + \int \rho(\mathbf{r})|\boldsymbol{\nu}(\mathbf{r})|^2\, C(\rho)|_{\rho=\rho(\mathbf{r})}d\mathbf{r}, \tag{32}$$

where $E_c[\rho]$ is the correlation energy functional of the conventional DFT. The expansion coefficient $C(\rho)$ has been devised so that Eq. (32) satisfies sum rules and bounds as much as one can. Our best expression of $C(\rho)$ is given by[29]

$$C(\rho) = \bar{C}_0 \frac{\hbar^2}{a_H^3 \varepsilon_H} \frac{e^{-\bar{\alpha} a_H^3 \rho} \rho^2}{(\rho - \bar{\delta}/a_H^3)^3}, \tag{33}$$

where $\bar{C}_0$, $\bar{\alpha}$, and $\bar{\delta}$ are dimensionless constants. If we choose $\bar{C}_0 < 0$, $\bar{\alpha} > 0$, and $0 < \bar{\delta} \ll a_H^3 \rho$, then all sum rules and bounds which are listed in Table I of Ref. 29 are satisfied with Eqs. (32) and (33).

The validity of the above VEA formulas has already been confirmed by comparing them with the exchange and correlation energies of the homogeneous electron liquid under a uniform magnetic field.[29,37]

B. Does the VEA formula satisfy Levy's asymptotic bound?

Here, we shall confirm that the above-mentioned VEA formula of the correlation energy satisfies Levy's asymptotic bound of the CDFT, i.e., Eq. (28). For the scaled basic variables, Eq. (32) is written as

$$\bar{E}_c[\rho_\lambda, \boldsymbol{\nu}_\lambda] = E_c[\rho_\lambda] + \int \rho(\mathbf{r}) |\boldsymbol{\nu}(\mathbf{r})|^2 \lambda^4 C[\lambda^3 \rho(\mathbf{r})] d\mathbf{r}, \tag{34}$$

where we use Eqs. (12) and (29) and the transformation of the integration variables. Taking the limit $\lambda \to \infty$ on both sides, we have

$$\lim_{\lambda\to\infty} \bar{E}_c[\rho_\lambda, \boldsymbol{\nu}_\lambda] = \lim_{\lambda\to\infty} E_c[\rho_\lambda] + \int \rho(\mathbf{r}) |\boldsymbol{\nu}(\mathbf{r})|^2 \{\lim_{\lambda\to\infty} \lambda^4 C[\lambda^3 \rho(\mathbf{r})]\} d\mathbf{r}. \tag{35}$$

The first term on the right-hand side has been already given by Levy.[33,34] That is,

$$\lim_{\lambda\to\infty} E_c[\rho_\lambda] = \text{const}. \tag{36}$$

Therefore, in order to satisfy Eq. (28), the following relation has to hold:

$$\int \rho(\mathbf{r}) |\boldsymbol{\nu}(\mathbf{r})|^2 \{\lim_{\lambda\to\infty} \lambda^4 C[\lambda^3 \rho(\mathbf{r})]\} d\mathbf{r} = 0 \quad \text{or const}. \tag{37}$$

As the sufficient condition for Eq. (37), we have

$$\lim_{\lambda\to\infty} \lambda^4 C(\lambda^3 \rho) = 0 \quad \text{or a positive power of } \rho. \tag{38}$$

Using Eq. (33), the left-hand side of Eq. (38) is given by

$$\lim_{\lambda\to\infty} \lambda^4 C(\lambda^3 \rho) = \lim_{\lambda\to\infty} \lambda^4 \frac{\hbar^2}{a_H^3 \varepsilon_H} \frac{\bar{C}_0 e^{-\bar{\alpha} a_H^3 \lambda^3 \rho} \lambda^6 \rho^2}{(\lambda^3 \rho - \bar{\delta}/a_H^3)^3} = \lim_{\lambda\to\infty} \frac{\hbar^2}{a_H^3 \varepsilon_H} \frac{\bar{C}_0 e^{-\bar{\alpha} a_H^3 \lambda^3 \rho} \lambda \rho^2}{[\rho - \bar{\delta}/(a_H \lambda)^3]^3} = 0. \tag{39}$$

In the third line, we use the fact that $\bar{\alpha}$ is positive. Therefore, it is shown that the VEA formula of the correlation energy satisfies Levy's asymptotic bound of the CDFT.

IV. EVALUATION OF THE LDA FORMULA

In this section, we shall show that Levy's asymptotic bound is *not* satisfied with the LDA of the CDFT. For this purpose, we shall use the correlation energy of a homogeneous electron liquid under a uniform magnetic field, which has been calculated by Skudlarski and Vignale within the random-phase approximation.[38] In their paper, the correlation energy per particle is given as a function of both the density parameter r_s and the occupation factor of the lowest Landau subband η. They gave the following approximate forms:[38]

$$\varepsilon_c^{\text{homo}}(r_s, \eta) = \begin{cases} \varepsilon_c^{\text{homo}}(r_s) + \Delta\varepsilon_c^{\text{homo}}(1, \eta)/r_s & \text{for } \eta > 0.1 \\ -1.606 r_s^{-3/4}(1 - 1.009\eta^{-1/4}) & \text{for } \eta < 0.3, \end{cases} \tag{40}$$

where $\varepsilon_c^{\text{homo}}(r_s, \eta)$ and $\varepsilon_c^{\text{homo}}(r_s)$ stand for the correlation energies per particle of the homogeneous electron liquid with and without a magnetic field, respectively. $\Delta\varepsilon_c^{\text{homo}}(1, \eta)$ is the difference between $\varepsilon_c^{\text{homo}}(r_s, \eta)$ and $\varepsilon_c^{\text{homo}}(r_s)$ at $r_s = 1$.

Using Eq. (40), the LDA of the CDFT can be defined by[22]

$$\bar{E}_c^{LDA}[\rho, \boldsymbol{\nu}] = \int \rho(\mathbf{r}) \varepsilon_c^{\text{homo}}[r_s(\mathbf{r}), \eta(\mathbf{r})] d\mathbf{r}. \tag{41}$$

Note that the left-hand side of Eq. (41) is explicitly written as a functional of $\rho(\mathbf{r})$ and $\boldsymbol{\nu}(\mathbf{r})$. This is because ρ, r_s, η, and $\boldsymbol{\nu}$ are related to each other by[38]

$$\rho = \left(\frac{3}{4\pi a_H^3 r_s^3} \right) \tag{42}$$

and

$$\rho = \frac{(\hbar/\varepsilon_H)^{3/2}}{2\pi^2 a_H^3} |\boldsymbol{\nu}|^{3/2} \sum_{N=0}^{\text{int}\{\eta^2\}} \sqrt{\eta^2 - N}, \tag{43}$$

where the symbol int$\{\eta^2\}$ means the integer part of η^2. Due to Eqs. (42) and (43), $\varepsilon_c^{\text{homo}}(r_s, \eta)$ is also regarded as a function of both ρ and $\boldsymbol{\nu}$.

If the density parameter and occupation factor which are determined by $\rho_\lambda(\mathbf{r})$ and $\boldsymbol{\nu}_\lambda(\mathbf{r})$ via Eqs. (42) and (43) are denoted as $r_{s\lambda}(\mathbf{r})$ and $\eta_\lambda(\mathbf{r})$, respectively, then Eq. (41) leads to

$$\bar{E}_c^{LDA}[\rho_\lambda, \boldsymbol{\nu}_\lambda] = \int \rho_\lambda(\mathbf{r}) \varepsilon_c^{\text{homo}}[r_{s\lambda}(\mathbf{r}), \eta_\lambda(\mathbf{r})] d\mathbf{r}. \tag{44}$$

Using Eqs. (12), (29), (42), and (43), $r_{s\lambda}(\mathbf{r})$ and $\eta_\lambda(\mathbf{r})$ are given by

$$r_{s\lambda}(\mathbf{r}) = \lambda^{-1} r_s(\mathbf{r}) \tag{45}$$

and

$$\eta_\lambda(\mathbf{r}) = \eta(\lambda \mathbf{r}), \tag{46}$$

respectively. The proof of Eq. (46) is given in Appendix B. Substituting Eqs. (45) and (46) into Eq. (44) and transforming the integration variables in Eq. (44), we get

$$\bar{E}_c^{LDA}[\rho_\lambda, \boldsymbol{\nu}_\lambda] = \int \rho(\mathbf{r}) \varepsilon_c^{\text{homo}}[\lambda^{-1} r_s(\mathbf{r}), \eta(\mathbf{r})] d\mathbf{r}. \quad (47)$$

By using Eq. (40) and the fact that $\varepsilon_c^{\text{homo}}(\lambda^{-1} r_s)$ causes the logarithmic divergence as λ goes infinity, it is confirmed that $\bar{E}_c^{LDA}[\rho_\lambda, \boldsymbol{\nu}_\lambda]$ becomes positive or negative infinity in the limit of $\lambda \to \infty$. That is to say, we have

$$\lim_{\lambda \to \infty} \bar{E}_c^{LDA}[\rho_\lambda, \boldsymbol{\nu}_\lambda] \neq \text{const.} \quad (48)$$

Consequently, Levy's asymptotic bound is not satisfied with the LDA of the CDFT.

V. CONCLUSION

An extension of Levy's asymptotic bound to the CDFT has been derived by using the coordinate scaling. This bound will provide some guideline in developing the correlation energy functional of the CDFT. We also evaluated both the VEA (Ref. 29) and LDA formulas of the CDFT by means of such a bound. It is found that the VEA formula exactly satisfies it, though the LDA does not. This means the validity of the VEA formula.

ACKNOWLEDGMENT

This work was partially supported by a Grant-in-Aid for Scientific Research in Priority Areas "Development of New Quantum Simulators and Quantum Design" of The Ministry of Education, Culture, Sports, Science, and Technology, Japan.

APPENDIX A: THE GROUND-STATE WAVE FUNCTION OF $\hat{H}_\lambda$

Suppose that $\Psi_\lambda(\mathbf{r}_1, \ldots, \mathbf{r}_N)$ and ε_λ are the ground-state wave function of $\hat{H}_\lambda$ and its eigenvalue, respectively, i.e.,

$$\hat{H}_\lambda \Psi_\lambda(\mathbf{r}_1, \ldots, \mathbf{r}_N) = \varepsilon_\lambda \Psi_\lambda(\mathbf{r}_1, \ldots, \mathbf{r}_N). \quad (A1)$$

If we assume that $\lambda^{3N/2} \Psi_s^{\lambda^{-1}}(\lambda \mathbf{r}_1, \ldots, \lambda \mathbf{r}_N)$ is not the ground-state wave function of $\hat{H}_\lambda$, then the corresponding eigenvalue $\lambda^2 E_s^{\lambda^{-1}}$ is larger than ε_λ,

$$\varepsilon_\lambda < \lambda^2 E_s^{\lambda^{-1}}. \quad (A2)$$

On the other hand, transforming Eq. (A1) by the coordinate scaling of electrons such that $\mathbf{r}_i \to \lambda \mathbf{r}_i$, we get

$$\left[\hat{T} + \int v_s(\mathbf{r}) \hat{\rho}(\mathbf{r}) d\mathbf{r} + \int \mathbf{A}_s(\mathbf{r}) \cdot \hat{\mathbf{j}}_p(\mathbf{r}) d\mathbf{r} + \lambda^{-1} \hat{W} \right] \times [\lambda^{-3N/2} \Psi_\lambda(\lambda^{-1} \mathbf{r}_1, \ldots, \lambda^{-1} \mathbf{r}_N)] = \lambda^{-2} \varepsilon_\lambda [\lambda^{-3N/2} \Psi_\lambda(\lambda^{-1} \mathbf{r}_1, \ldots, \lambda^{-1} \mathbf{r}_N)]. \quad (A3)$$

Substituting $\alpha = \lambda^{-1}$ into Eq. (8), we also obtain

$$\left[\hat{T} + \int v_s(\mathbf{r}) \hat{\rho}(\mathbf{r}) d\mathbf{r} + \int \mathbf{A}_s(\mathbf{r}) \cdot \hat{\mathbf{j}}_p(\mathbf{r}) d\mathbf{r} + \lambda^{-1} \hat{W} \right] \times \Psi_s^{\lambda^{-1}}(\mathbf{r}_1, \ldots, \mathbf{r}_N) = E_s^{\lambda^{-1}} \Psi_s^{\lambda^{-1}}(\mathbf{r}_1, \ldots, \mathbf{r}_N). \quad (A4)$$

Comparing Eqs. (A3) and (A4) and taking into account the fact that $E_s^{\lambda^{-1}}$ is the ground-state energy of Eq. (A4), we obtain the inequality

$$\lambda^{-2} \varepsilon_\lambda \geqslant E_s^{\lambda^{-1}}. \quad (A5)$$

This inequality is inconsistent with Eq. (A2). Consequently, $\lambda^2 E_s^{\lambda^{-1}}$ is the ground-state energy of $\hat{H}_\lambda$, and the corresponding wave function $\lambda^{3N/2} \Psi_s^{\lambda^{-1}}(\lambda \mathbf{r}_1, \ldots, \lambda \mathbf{r}_N)$ is the ground-state wave function of $\hat{H}_\lambda$.

APPENDIX B: PROOF OF EQ. (46)

Substitution of Eqs. (12) and (29) into Eq. (43) leads to

$$\rho(\lambda \mathbf{r}) = \frac{(\hbar/\varepsilon_H)^{3/2}}{2\pi^2 a_H^3} |\boldsymbol{\nu}(\lambda \mathbf{r})|^{3/2} \sum_{N=0}^{\text{int}\{\eta_\lambda(\mathbf{r})\}} \sqrt{\eta_\lambda^2(\mathbf{r}) - N}, \quad (B1)$$

where $\eta_\lambda(\mathbf{r})$ denotes the occupation factor which are determined by $\rho_\lambda(\mathbf{r})$ and $\boldsymbol{\nu}_\lambda(\mathbf{r})$ via Eq. (43). On the other hand, we have

$$\rho(\lambda \mathbf{r}) = \frac{(\hbar/\varepsilon_H)^{3/2}}{2\pi^2 a_H^3} |\boldsymbol{\nu}(\lambda \mathbf{r})|^{3/2} \sum_{N=0}^{\text{int}\{\eta_\lambda(\mathbf{r})\}} \sqrt{\eta^2(\lambda \mathbf{r}) - N}. \quad (B2)$$

From Eqs. (B1) and (B2), we get the equality

$$\sum_{N=0}^{\text{int}\{\eta(\lambda \mathbf{r})\}} \sqrt{\eta^2(\lambda \mathbf{r}) - N} = \sum_{N=0}^{\text{int}\{\eta_\lambda(\mathbf{r})\}} \sqrt{\eta_\lambda^2(\mathbf{r}) - N}. \quad (B3)$$

Taking into the account the fact that the function which is defined by $f(x) = \Sigma_{N=0}^{\text{int}\{x\}} \sqrt{x^2 - N}$ is a monotonically increasing function, Eq. (B3) leads to $\eta_\lambda(\mathbf{r}) = \eta(\lambda \mathbf{r})$.

[1] P. Hohenberg and W. Kohn, Phys. Rev. **136**, B864 (1964).
[2] W. Kohn and L. J. Sham, Phys. Rev. **140**, A1133 (1965).
[3] M. Higuchi and K. Higuchi, Phys. Rev. B **69**, 035113 (2004).
[4] K. Higuchi and M. Higuchi, Phys. Rev. B **69**, 165118 (2004).
[5] Á. Nagy, S. Liu, and L. Bartolloti, J. Chem. Phys. **122**, 134107 (2005).
[6] U. von Barth and L. Hedin, J. Phys. C **5**, 1629 (1972).
[7] M. M. Pant and A. K. Rajagopal, Solid State Commun. **10**, 1157 (1972).
[8] G. Vignale and M. Rasolt, Phys. Rev. Lett. **59**, 2360 (1987).
[9] G. Vignale and M. Rasolt, Phys. Rev. B **37**, 10685 (1988).
[10] M. Higuchi and A. Hasegawa, J. Phys. Soc. Jpn. **66**, 149 (1997).
[11] M. Higuchi and A. Hasegawa, J. Phys. Soc. Jpn. **67**, 2037 (1998).
[12] V. I. Anisimov, J. Zaanen, and O. K. Andersen, Phys. Rev. B **44**,

943 (1991).
[13] V. I. Anisimov and O. Gunnarsson, Phys. Rev. B **43**, 7570 (1991).
[14] V. I. Anisimov, I. V. Solovyev, M. A. Korotin, M. T. Czyzyk, and G. A. Sawatzky, Phys. Rev. B **48**, 16929 (1993).
[15] A. I. Liechtenstein, V. I. Anisimov, and J. Zaanen, Phys. Rev. B **52**, R5467 (1995).
[16] G. Vignale, M. Rasolt, and D. J. W. Geldart, Phys. Rev. B **37**, 2502 (1988).
[17] G. Vignale, M. Rasolt, and D. J. W. Geldart, Adv. Quantum Chem. **21**, 235 (1990).
[18] S. Erhard and E. K. U. Gross, Phys. Rev. A **53**, R5 (1996).
[19] S. Liu, Phys. Rev. A **54**, 1328 (1996).
[20] K. Capelle and E. K. U. Gross, Phys. Rev. Lett. **78**, 1872 (1997).
[21] A. M. Lee and N. C. Handy, Phys. Rev. A **59**, 209 (1999).
[22] M. Higuchi and K. Higuchi, Phys. Rev. B **65**, 195122 (2002).
[23] A. D. Becke, J. Chem. Phys. **117**, 6935 (2002).
[24] S. N. Maximoff, M. Ernzerhof, and G. E. Scuseria, J. Chem. Phys. **120**, 2105 (2004).
[25] K. Higuchi and M. Higuchi, Phys. Rev. B **71**, 035116 (2005).
[26] W. Zhu and S. B. Trickey, Phys. Rev. A **72**, 022501 (2005).
[27] J. Tao and J. P. Perdew, Phys. Rev. Lett. **95**, 196403 (2005).
[28] Á. Nagy, Int. J. Quantum Chem. **106**, 1043 (2006).
[29] K. Higuchi and M. Higuchi, Phys. Rev. B **74**, 195122 (2006).
[30] J. P. Perdew and Y. Wang, Phys. Rev. B **33**, 8800 (1986).
[31] J. P. Perdew, in *Electronic Structure of Solids '91*, edited by P. Ziesche and H. Eschrig (Akademie, Berlin, 1991), p. 11.
[32] J. P. Perdew, K. Burke, and M. Ernzerhof, Phys. Rev. Lett. **77**, 3865 (1996).
[33] M. Levy, Int. J. Quantum Chem. **S23**, 617 (1989).
[34] M. Levy, Phys. Rev. A **43**, 4637 (1991).
[35] D. Pines, *Solid State Physics* (Academic, New York, 1957), Vol. 1, p. 367.
[36] P. Noziéres and D. Pines, Phys. Rev. **111**, 442 (1958).
[37] Y. Takada and H. Goto, J. Phys.: Condens. Matter **10**, 11315 (1998).
[38] P. Skudlarski and G. Vignale, Phys. Rev. B **47**, 16647 (1993).